

Sulfur dioxide emissions in Iran and environmental impacts of sulfur recovery plant in Tabriz Oil Refinery

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Abstract

Background: Combustion of fossil fuels contributes to sulfur dioxide (SO₂) emissions. To deal with this issue, the government of Iran has appointed the oil refineries to upgrade their installations and produce high quality fuels. Thus, this study investigated the status of SO₂ emissions in Iran and the capability of advanced technologies to control SO₂ emissions.

Methods: The status of SO₂ emissions was reviewed and discussed through national online reports. Meanwhile, the environmental impacts of sulfur recovery and tail gas treatment (TGT) plant (STP) were assessed by applying rapid impact assessment matrix (RIAM) for implementation and non-implementation alternatives in Tabriz Oil Refinery Company (TORC).

Results: SO₂ emissions have been increased by 2.1 times during 2004-2014 in Iran. Power plants and transportation play a significant role in this regard and overall contribute 82% of emissions. Among the other fossil fuels, fuel oil and gasoil account for 95% of SO₂ emissions. Based on the environmental impact assessments (EIAs), sulfur recovery management and enhancing sulfur removal efficiency from flue gas up to 99.9% are two main positive environmental aspects of STP project that would enable TORC to prevent 87 600 tons of SO₂ emissions, annually. Nevertheless, flue gas and sour gas streams which have been determined as probable pollution sources of process, should be managed through proper monitoring framework.

Conclusion: The increasing trend of SO₂ emissions and significant role of fuel oil and gasoil has required Iranian oil refineries to enhance the quality of fuels by employing clean and cost-effective technologies.

Keywords: Air pollution, Fossil fuels, Oil and gas industry, Environmental assessment, Tabriz

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Introduction

Sulfur dioxide (SO₂), emitting from either natural or anthropogenic sources, is one of the important air pollutants leading to acid precipitations, climatic changes and health problems. Previous studies showed that short- or long-term exposure to SO₂ contributes to increase of hospital admissions and mortality due to respiratory and cardiac problems including chronic obstructive pulmonary disease and acute myocardial infraction (1-3). More than 70% of global SO₂ emissions is accounted for anthropogenic sources including power plants, refineries and vehicles (4). Fioletov et al detected 491 emitting sources via ozone monitoring instrument and suggested that the oil and gas industries are the second anthropogenic sources of SO₂ emissions over the globe (5). Sulfur oxides along with other pollutants such as

nitrogen oxides (NO_x), volatile organic compounds (VOCs) and carbon monoxide (CO) are the group of pollutants generated from the operations of oil industries. Sulfur content of crude oil varies from 0.05 to 6 by weight percentage, approximately (6).

The investigations indicate that global SO₂ emission has followed an increasing trend during 2000 to 2005 and has been resulted from industrial activities development in Asian countries, particularly in China and India. Despite the fact that flue gas desulfurization (FGD) and some other practical measures in power plants of China, the United States and Europe have resulted in great decline of emissions rate since 2006 (7-10), the emissions from fuel combustion have remained constant since 1980 (10,11). To achieve health benefits from improved outdoor air quality (12), sulfur-limit transportation fuels like diesel oil



and gasoline, have been gradually restricted, in Germany. For instance, diesel fuel standards have been decreased from 5000 to 350 ppm while in the United States, it has been changed from 500 in 1993 to 15 ppm since 2006 (13). In this regard, the government of Iran has appointed oil refinery companies to upgrade their installations and provide high quality fuels by the end of 2019. These stringent standards require gas plants and crude oil refineries to employ reliable and cost-effective technologies for advanced sulfur recovery (14,15). Among biodesulfurization, surface adsorption and oxidation, hydrodesulfurization (HDS) is the most common process applied to eliminate sulfur from crude oil in petroleum industry. In this process, sulfur containing compounds react with hydrogen on $\text{Mo}/\text{Al}_2\text{O}_3\text{-Co}$ or $\text{Mo}/\text{Al}_2\text{O}_3\text{-Ni}$ catalysts to yield hydrocarbons and H_2S (16).

East Azerbaijan province with a population of 3 724 620 people had gasoil consumption of 950 million liters in 2015 and have ranked 23rd in the country (17). Tabriz Oil Refinery Company (TORC) is located in the northwest of Iran, near the city of Tabriz (Figure 1). This refinery has a nominal capacity of 110 000 barrels per day producing 6 types of fuels including gasoil, fuel oil, gasoline, liquid petroleum gas (LPG) and kerosene (18). Currently, 35.5 by weight percentage of refined crude oil is converted to

gasoil in TORC. Sulfur content of crude oil needed to be reduced from 150 to ≤ 50 ppm to provide Euro 4 gasoil, utilizing gasoil hydrotreating plant (GHP). This process would enable TORC to produce 4.77 million liters Euro 4 gasoil daily, comprising 19.87% of the total gasoil produced in the country. Nevertheless, the prospective sour gas streams of this plant would impose exceeded burden on the sulfur recovery unit (SRU).

To address this problem, employing sulfur treatment plant (STP) technology is expected to bring about the following advantages:

- 1) Raise liquid sulfur recovery capacity from 80 up to 190 tons per day
- 2) Raise sulfur removal efficiency from sour and acid gas streams up to 99.9%
- 3) Minimize hydrogen sulfur (H_2S) emissions from flue gas

Figure 2 describes the procedure of sulfur recovery process in STP; the sour and acid gas streams from Amine treatment unit (ATU), sour water stripping unit (SWSU) and GHP are collected and sent to SRU with clause process. Claus process is a main technique for processing high amount of acid streams and involves two reactions; initially, SO_2 is generated through the combustion of H_2S using air or O_2 , thereafter, the reaction of SO_2 with H_2S will

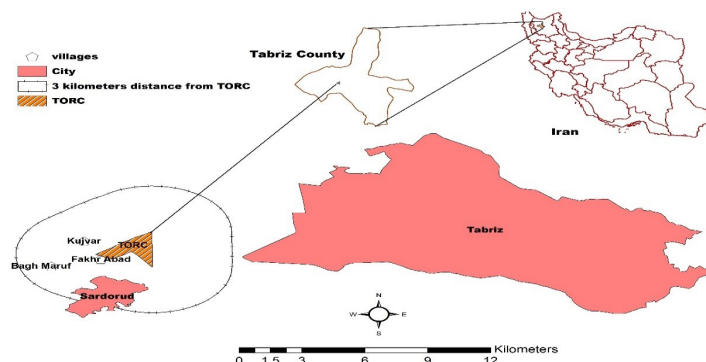


Figure 1. Location of Tabriz Oil Refinery Company.

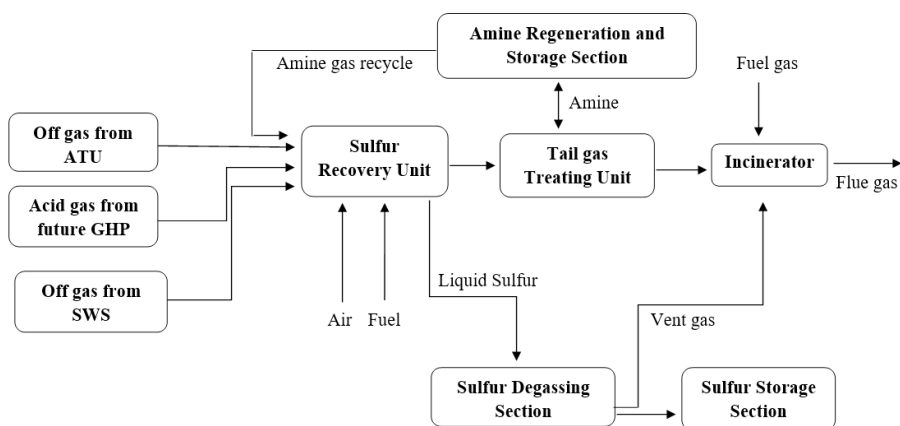


Figure 2. Sulfur recovery and tail gas treating plant diagram.

produce liquid sulfur which would finally store in solid form. Stream of sulfur oxides (H_2S , CS_2 , SO_2 and COS) generated through Claus process would sent to tail gas treatment plant (TGT). Incorporation of high temperature and hydrogen in advanced treatment, will convert these compounds to H_2S gas. After absorption of H_2S by amine solution, treated tail gas will be finally sent to incinerator (19,20).

Based on the national environmental regulations, all industrial plants are subjected to environmental impact assessment (EIA). EIA is defined as the systematic identification and evaluation of potential impacts of proposed projects, plans, programs, or legislative actions relative to the physical, chemical, biological, cultural, and socio-economic components of the total environment. EIA is an environmental management tool aiming at identifying environmental issues and providing solutions to prevent or minimize these problems to desired levels. It also provides an environmental management plan, which includes a monitoring program. Rapid impact assessment matrix (RIAM) is one of the useful impact assessment tools that has been employed in many case studies for various purposes including landfills, water management, etc. (21-23).

Present study was conducted to provide an overview of SO_2 emissions in Iran through online national reports and assess the environmental impact of STP process by applying RIAM method. Indeed, the overall approach of this paper was to investigate the participation rate of different sectors and fuels in SO_2 emissions and also the capability of advanced technologies to control emissions.

Methods

SO_2 emissions in Iran

To provide an overview of SO_2 emissions status in Iran, the latest online documents published by the Ministry of Energy were investigated. The trend of SO_2 emissions was reviewed during 2001 to 2014, whilst the contribution of sources has been investigated during 2014. Findings provided us with both SO_2 emissions trend and share of different energy sectors and fuels in this regard.

RIAM description for STP

RIAM provides a transparent and permanent record of the analysis process while at the same time organizing the EIA procedure, which in turn reduces the time spent on executing EIAs. The simple structured form of RIAM allows reanalysis and in-depth analysis of the selected components in a rapid and precise manner. In addition, the capability of comparing different options makes this method flexible which provides decision makers with transparent judgments (24).

RIAM is based on the standard definition of assessment criteria including importance, magnitude, permanence, reversibility and cumulative. Each criterion involves a range of values with specific description (Table 1).

Table 1. Assessment criteria of rapid impact assessment matrix

| Criteria | Scale | Description |
|----------|-------|---------------------------------------|
| A_1 | 0 | No importance |
| | 1 | Important only to the local condition |
| | 2 | Important to the areas immediately |
| | 3 | Important to national interests |
| | 4 | Important to international interest |
| A_2 | +3 | Major positive benefits |
| | +2 | Significant improvement in status quo |
| | +1 | Improvement in status quo |
| | 0 | No change |
| | -1 | Negative change to status quo |
| B_1 | -2 | Significant negative impacts |
| | -3 | Major negative impacts |
| | 1 | No change |
| | 2 | Temporary |
| | 3 | Permanent |
| B_2 | 1 | No change |
| | 2 | Reversible |
| | 3 | Irreversible |
| B_3 | 1 | No change |
| | 2 | Non-cumulative |
| | 3 | Cumulative |

The values are selected based on the overall predicted impacts of activities on the environmental components. Thereafter, the selected values will undergo the following mathematical calculations (24):

$$A1 \times A2 = aT$$

$$B1 + B2 + B3 = bT$$

$$aT \times bT = ES$$

The scores of importance (A_1) and magnitude (A_2) are the criteria that can individually change the score, so the application of a multiplier ensures that the weight of each score is counted. While, the scores of permanence (B_1), reversibility (B_2) and cumulative (B_3) are added together to ensure that the individual score wouldn't affect the obtained score (bT). Multiplying (aT) and (bT) scores together, will provide a final environmental score (ES) for each environmental component (24). To provide a more certain assessment, ES values will be set into one of the range bands described in Table 2. In this study, the scores of assessment criteria were estimated based on the location, scale and the nature of desulphurization process of STP and assessment carried out according to the range bands. Finally, all positive and negative impacts were presented in graphical form and discussed (24).

STP would operate in an area of about 0.4 hectares beyond the existing sulfur recovery plant. This location is sand-filled and devoid of any vegetation or special habitat. Meanwhile, most of the required facilities including access roads, energy sources, and wastewater management system are available in place. Hence, the impact assessment was conducted for implementation (construction and

Table 2. Conversion of environmental scores to range bands

| ES | RB | Description |
|-------------|----|------------------------------|
| 72 to 108 | E | Major positive impacts |
| 36 to 71 | D | Significant positive impacts |
| 19 to 35 | C | Moderately positive impacts |
| 10 to 18 | B | Positive impacts |
| 1 to 9 | A | Slightly positive impacts |
| 0 | N | No change |
| -1 to -9 | -A | Slightly negative impacts |
| -10 to -18 | -B | Negative impacts |
| -19 to -35 | -C | Moderately negative impacts |
| -36 to -71 | -D | Significant negative impacts |
| -72 to -108 | -E | Major negative impacts |

Abbreviations: RB, range bands; ES, environmental score.

operation phases) and non-implementation alternatives separately based on the following assumptions:

1. The prospective sour gas stream from GHP will impose additional burden on SRU in case of non-implementation alternatives
2. STP technology is an environmentally friendly approach which deals with the additional sour gas streams with minimum pollution in case of implementation alternatives

The impacts of STP were evaluated against 15 various environmental components. Environmental impacts were categorized into 4 groups including physical, biological, cultural and socioeconomic environments. Physical components focus on the changes in the quality and quantity of water sources and air pollutants. Biological components refer to the existed fauna and flora, while the cultural and socioeconomic components are mainly concerned with human life quality.

Results

General trend of SO₂ emissions is represented in Figure 3 (25), which was positive during 2001-2014; emission rate has started to increase from 2004 and after a steep slope between 2006-2007, it has reached the highest level of 678078 tons in 2009 and after a rapid decrease in 2010, it started to increase with a mild slope up until 2013, however, the emission rate was partially decreased

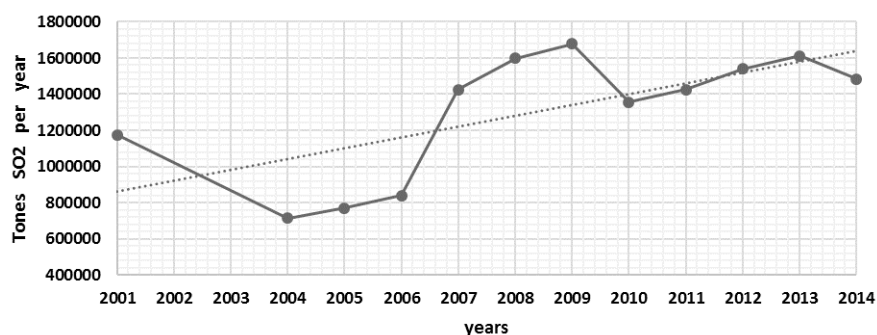


Figure 3. Trend of SO₂ emissions from all energy-consuming sectors including power plants, refineries, transportation, agriculture, etc. during 2001-2014 in Iran

about 130,246 tons by 2014. Figure 4 indicates the share of various sectors in SO₂ emissions as follows: Power plants > transportation > industry > agriculture > (public, domestic, commercial). Power plants and transportation overall contribute 82% of the emissions. Fuel oil together with gasoil account for 95% of SO₂ emissions, while the remained 5 % belongs to gasoline, heavy jet oil (ATK), kerosene and natural gas, totally (Figure 5).

Table 3 and Figure 6 represent the final yields of the assessment for both non-implementation and implementation alternatives. The results of assessment showed that non-implementation alternatives kept 86 % of the environment safe and stable. Nevertheless, incomplete treatment and discharge of prospective additional sour gas stream from GHP would pose negative impact on air quality. Surface water pollution is another undesired impact caused by solid sulfur washing.

The project implementation alternatives did not have any positive or negative impact on 70 % of the environment. 16% of the entire impacts of implementation alternatives were described as slightly negative impacts (-A) which mainly resulted from construction activities including embankment, excavation and transportation of construction materials or staff. These activities affect soil, air and surface water quality as well as road traffic. However, they are predicted to be temporary and

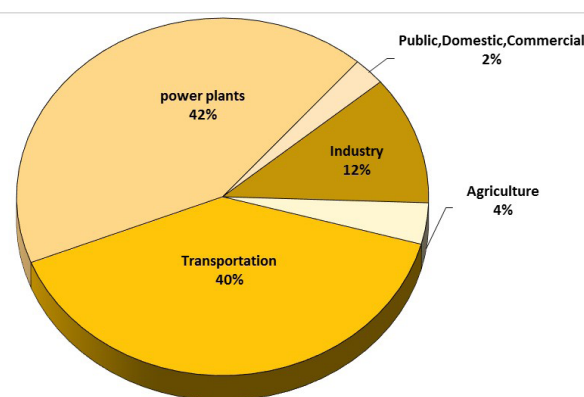


Figure 4. Share of different sectors in SO₂ emissions during 2014 in Iran.

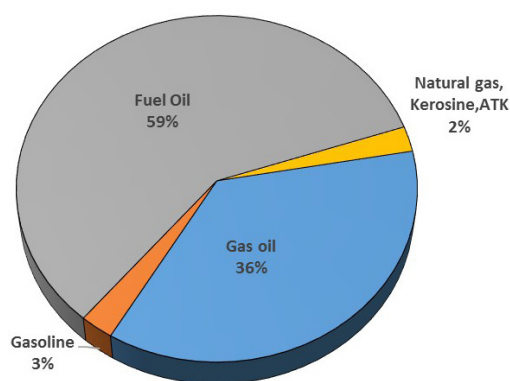


Figure 5. Share of different fuels in SO₂ emissions during 2014 in Iran

reversible. The remained 14% of positive impacts are completely associated with air quality; employment of novel technology for advanced treatment of tail gas and SO₂ which will be able to convert the ES of air quality from -28 to 30. Besides, providing job opportunity and industrial application of land were classified as slightly positive impacts (A) of STP employment. The overall identified impacts of this project were concerned with physical and socioeconomic environments. However, biological and cultural environments were not affected by the implementation or non-implementation alternatives.

Discussion

SO₂ emissions

Results of the present study revealed that SO₂ emissions followed a growing trend during 2001-2014 (Figure 3). Among all energy consuming sectors, power plants and transportation have more contribution to emissions, respectively. However, compared to previous years, the share of power plants (42%) was almost equal to that of transportation (40%) in 2014, which probably implies the

alternative utilization of clean fuel in power industries (Figure 4).

Generally, natural gas, fuel oil and gasoil are the main sources of energy for electricity generation in Iran's power plants. According to Mazandarani et al studies, consumption of natural gas, diesel and fuel oil have been respectively increased by 18.6, 3.1 and 8.4 times in power plants during 1979-2008 (26) and consequently, given the eleven-time increase in electricity consumption, nominal capacity reached up to 56181 MW by 2009 and more than 96 % of electricity has produced by fossil fuel-based power plants during the year (27). This report is greatly in line with general trend of SO₂ emissions (Figure 3) and interestingly expresses the peak of emission rates in 2009. Actually, the economic growth of Iran depends on oil and gas industries. However, beyond the supply side of the energy, the problem of pollutant emissions over local and global atmosphere should be addressed (28). As Figure 5 indicates, fuel oil and gasoil account for 82% of SO₂ emissions in 2014. On the other hand, gasoil (diesel fuel) and gasoline are two important fuels in transportation sector but gasoline has negligible share in SO₂ emissions compared to gasoil (Figure 5). Consequently, due to its extensive utilization both in power industry and transportation, the sulfur content of diesel fuels can be considered as a key factor of fuel standards (29).

It is well known that sulfur content of fuels plays a significant role in SO₂ emissions; Nazari et al reported that fuel oils consumed in Iranian steam power plants contain an average of 2.8 weight percentage of sulfur which makes their SO₂ emission factors to be 4 and 7 times higher than that of gas turbines and combined cycle power plants, respectively (30). Relevantly, Tan et al suggested that physical and chemical properties of diesel fuels have important influence on exhaust emissions. SO₂ emission continuously increases along with the engine load, and

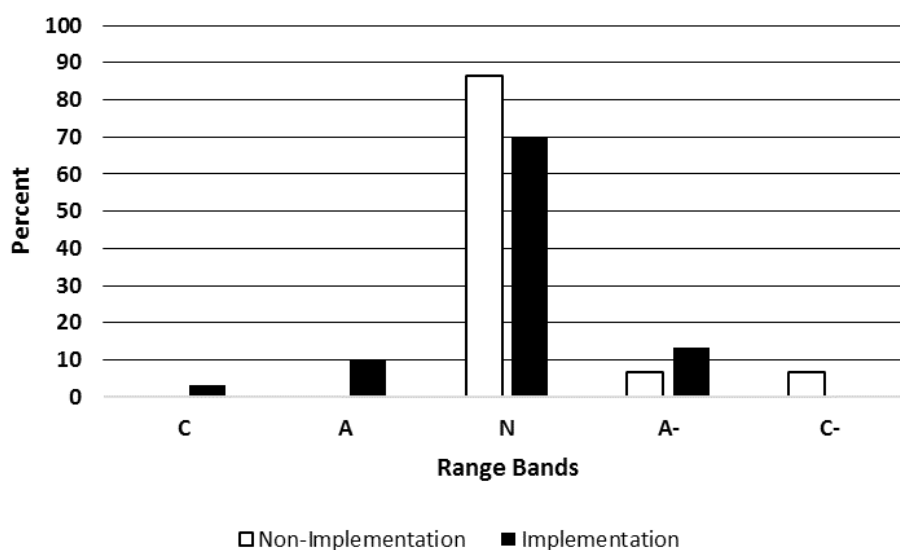


Figure 6. Comparison of Non-implementation and implementation alternatives for STP.

Table 3. Environmental impact assessment of STP for Non-implementation (NI), construction and operation

| Environmental Components | | | Phase | Criteria | | | | | |
|--------------------------|-------------------|--------------|-------|----------------|----------------|----------------|----------------|----------------|----|
| | | | | A ₁ | A ₂ | B ₁ | B ₂ | B ₃ | ES |
| Physical Environment | Climate | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | Air | NI | 2 | -2 | 3 | 1 | 3 | -28 | -C |
| | | Construction | 1 | -1 | 1 | 2 | 1 | -4 | -A |
| | | Operation | 3 | +2 | 3 | 1 | 1 | 30 | C |
| | Soil | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 1 | -1 | 3 | 3 | 1 | -7 | -A |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | Surface water | NI | 2 | -1 | 1 | 1 | 1 | -3 | -A |
| | | Construction | 2 | -1 | 1 | 1 | 1 | -6 | -A |
| | | Operation | 2 | -1 | 1 | 1 | 1 | -6 | -A |
| | Ground water | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| Biological Environment | Vegetation | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | Animals | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | Habitat | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | Protected areas | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| Socioeconomic | Employment | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 1 | +1 | 1 | 2 | 1 | 4 | A |
| | | Operation | 1 | +1 | 1 | 2 | 1 | 4 | A |
| | Land application | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 1 | +1 | 3 | 1 | 1 | 5 | A |
| | Road traffic | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 1 | -1 | 1 | 1 | 3 | -5 | -A |
| | | Operation | 0 | -1 | 2 | 1 | 3 | 0 | N |
| | Migration | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| Culture | Religious beliefs | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | Cultural heritage | NI | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Construction | 0 | 0 | 1 | 1 | 1 | 0 | N |
| | | Operation | 0 | 0 | 1 | 1 | 1 | 0 | N |

linearly varies with the fuel sulfur contents (31). Zhang et al also stated that sulfur content of crude oil fields and employment of advanced desulfurization technologies, were crucial factors in sulfur content of diesel and gasoline fuels in River delta regions (32).

Environmental impact assessment of STP

The overall analysis of EIA indicated that STP process is an environmentally friendly technology, aiming at enhancing sulfur removal efficiency from acid gas streams with

minimum emissions. Moreover, the advanced treatment of tail gas would enable the extension of sulfur recovery level to 200 tons per day which is equivalent to prevention of 240 tons of SO₂ emissions daily. The entire process will increase sulfur recovery capacity up to 439585 tons per year. On the other hand, undesirable impacts of STP project can be effectively managed based on the recommended mitigation measures mentioned in Table 4. Desulphurization facilities occupy a small part of the refinery, which, if properly utilized, they won't threaten

Table 4. Mitigation measures and management program for construction activities and operation system of STP

| Construction Phase | |
|---------------------------------|--|
| Air quality | Selecting roof and proper location for trash dumping Water spraying of trash in regular intervals to reduce dust release Observing the speed limit by drivers |
| Water quality | Constructing drainage canal for run-off control Dedicating construction activities during low-rain seasons |
| Noise pollution control | Selecting the proper equipment with minimum level of noise Regular maintenance and servicing of equipment and installations |
| Waste management | Meeting source reduction, source segregation, reuse and recycling principles Using proper waste containers Regular collection and disposal |
| Operation System Control | |
| Sulfur recovery and TGT section | The whole STP performance should meet the environmental agency standards for acceptable level of emissions. Minimum 96 % conversion at the end of run should be achieved within the entire Claus plant. All processes of advanced treatment including hydrogenation, quench section, filters, amine absorbing and regeneration section should be performed; SO ₂ leakage and lean amine temperature are two key factors of TGT section that should be controlled. Appropriate analyzers (H ₂ S and H ₂) for quench and absorber towers should be considered. Maximum CS ₂ and COS hydrolysis should be achieved in hydrogenation section. |
| Incineration section | Oxidation of SO ₂ should be completely done at proper temperature levels to reduce H ₂ S emissions to less than 10 ppm. Concentration of flue gas pollutants should be measured and monitored, continually. |
| Pipes and installations | Performance of installations and the pipes should be controlled and managed in order to prevent SO ₂ and H ₂ S emissions and solid sulfur leakage. |

public health of nearby residential areas, located within 3 km away from refinery (Figure 1). However, emissions of H₂S and SO₂ during the process should be repeatedly monitored through the environmental management system. This system assists the operators to achieve a safe operation based on the identification of pollution source and its magnitude; in case of STP, for instance, the flue gas of incinerator and release of sulfur gas from installations are considered as the main pollution sources. To control these pollutants, SO₂ and H₂S levels should be recorded applying fixed detectors near the furnace or reactors. Besides, the staff should be equipped with portable detectors to be aware of exceeded level of sulfur gases at working area.

Conclusion

Generally, Power plants are defined as the major contributors to SO₂ emissions. However, desulfurization of flue gas and progressive utilization of clean fuels in electricity industries over recent years, is going to draw attentions toward transportation fuels. Due to extensive utilization of gasoil in transportation and power plants, more emphasis should be placed on its quality. In this regard, Iranian oil refineries aim at producing Euro 4 gasoil, employing clean and cost-effective technologies. It is estimated that employing hydro-desulfurization combined with STP technology in TORC, will extend sulfur recovery capacity up to 73000 tons yearly which will subsequently result in preventing 87600 tons of SO₂ emissions, annually.

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Ethical issues

It is confirmed that this manuscript is an original work and all data collection and analysis have been carried out by the authors. Meanwhile, it has not been or won't be published elsewhere separately.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

All authors equally participated in data collection, analysis and reviewing the manuscript.

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